

<u>4.</u>	ENVIRONMENTAL	RADIOLOGICAL	PROGRAI	M INFORMATION	<u> </u>
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4.1. Description of Monitoring Program

The radioactivity of the environment around ANL-E in 1999 was determined by measuring radionuclide concentrations in naturally occurring materials and by measuring the external penetrating radiation dose. Sample collections and measurements were made at the site perimeter and off site for comparative purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results.

Because radioactivity is primarily transported by air and water, the sample collection program concentrates on these media. In addition, samples of materials from the streambeds also are analyzed. The program follows the guidance provided in the DOE Environmental Regulatory Guide. ¹⁰ The results of radioactivity measurements are expressed in terms of pCi/L for water; fCi/m³ and aCi/m³ for air; and pCi/g and fCi/g for bottom sediment. Penetrating radiation measurements are reported in units of mrem/yr, and population dose is reported in units of man-rems.

DOE has provided guidance¹¹ for effective dose equivalent calculations for members of the public based on International Commission on Radiological Protection (ICRP) Publications 26 and 30. 12,13 Those procedures have been used in preparing this report. The methodology requires that three components be calculated:(1) the committed effective dose equivalent (CEDE) from all sources of ingestion, (2) the CEDE from inhalation, and (3) the direct effective dose equivalent from external radiation. These three components were summed for comparison with the DOE effective dose equivalent limits for environmental exposure. The guidance requires that sufficient data on exposure to radionuclide sources be available to ensure that at least 90% of the total CEDE is accounted for. The primary radiation dose limit for members of the public is 100 mrem/yr. The effective dose equivalents for members of the public from all routine DOE operations, natural background and medical exposures excluded, shall not exceed 100 mrem/yr and must adhere to the as-low-as-reasonably-achievable (ALARA) process or be as far below the limits as is practical, taking into account social, economic, technical, practical, and public policy considerations. Routine DOE operations are normally planned operations and exclude actual or potential accidental or unplanned releases.

The measured or calculated environmental radionuclide concentrations were converted to a 50-year CEDE with the use of the CEDE factors¹⁴ and were compared with the annual dose limits for uncontrolled areas. The CEDEs were calculated from the DOE Derived Concentration Guides (DCGs)¹¹ for members of the public on the basis of a radiation dose of 100 mrem/yr. The numerical values of the CEDE factors used in this report are provided later in this chapter (Table 4.26). Although the CEDE factors apply only to concentrations above natural levels, for comparative purposes, the calculated dose is sometimes given in this report for radioactivities that are primarily of natural origin. Occasionally, other standards are used, and their sources are identified in the text.

4.2. Air

The radioactive content of particles in the air was determined by collecting and analyzing air filter samples. The sampling locations are shown in Figures 1.1 and 1.2. Separate collections were made for specific radiochemical analyses and for gross alpha, gross beta, and gamma-ray spectrometry. The latter measurements were taken from samples collected continuously on laminated glass fiber filters (changed weekly) at 12 locations at the ANL-E site perimeter by using PM_{10} units (particles less than 10 micrometers) and at 5 off-site locations.

Samples were collected at the site perimeter to determine whether a statistically significant difference exists between perimeter measurements and measurements taken from samples collected at various off-site locations. The off-site samples establish the local background concentrations of naturally occurring or ubiquitous man-made radionuclides, such as from nuclear weapons testing fallout. Higher levels of radioactivity in the air measured at the site perimeter may indicate radioactivity releases from ANL-E, provided that the perimeter samples are greater than the background samples by an amount greater than the relative error of the measurement. The relative error is a result of natural variation in background concentrations as well as sampling and measurement error. This relative error is typically 5 to 20% of the measurement value for most of the analyses, but approaches 100% at values near the detection limit of the instrument.

Table 4.1 summarizes the total alpha and beta activities in the individual weekly samples. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for a 0.30-MeV beta and a 5.5-MeV alpha on filter paper. The results were obtained by measuring the samples four days after they were collected to avoid counting the natural activity due to short-lived radon and thoron decay products. This activity is normally present in air and disappears within four days by radioactive decay. The average concentrations of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples, are given in Table 4.2. The gamma-ray detector is a shielded germanium diode calibrated for each gamma-ray—emitting nuclide measured.

The alpha activity, principally due to naturally occurring nuclides, averaged the same as in the past several years and was within its normal range. The perimeter beta activity averaged 20 fCi/m³, which is similar to the average value for the past five years. The gamma-ray emitters listed in Table 4.2 are those that have been present in the air for the past five years and are of natural origin. The beryllium-7 concentration increases in the spring, which indicates its stratospheric origin. The concentration of lead-210 in the air is due to the radioactive decay of gaseous radon-222 and is similar to the concentration last year.

The annual average alpha and beta activities since 1985 are displayed in Figure 4.1. The elevated beta activity in 1986 was due to fallout from the Chernobyl incident. If the radionuclides

TABLE 4.1

Total Alpha and Beta Activities in Air Filter Samples, 1999

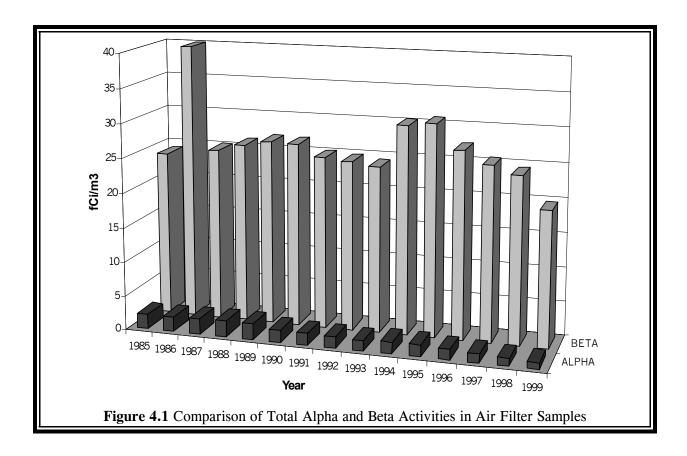
(concentrations in fCi/m³)

			Alpha Activity		Beta	Activity		
Month	Location	No. of Samples	Avg.	Min.	Max.	Avg.	Min.	Max.
January	Perimeter	25	0.8	0.1	1.7	23.7	11.6	41.7
Ž	Off-Site	19	1.7	0.5	3.5	29.2	16.7	57.6
February	Perimeter	44	0.7	0.2	1.1	19.0	9.6	27.6
	Off-Site	14	0.8	0.4	1.5	18.4	14.4	23.8
March	Perimeter	60	0.9	0.3	1.5	18.8	9.6	26.7
	Off-Site	17	1.1	0.7	1.6	16.2	11.3	26.8
April	Perimeter	46	0.8	0.2	1.5	15.4	7.0	23.7
	Off-Site	14	0.9	0.5	1.2	12.9	9.9	16.1
May	Perimeter	46	0.9	0.3	1.8	14.4	6.1	21.0
	Off-Site	16	0.9	0.3	2.1	12.6	8.2	24.5
June	Perimeter	60	0.7	0.1	1.6	15.0	5.6	30.3
	Off-Site	20	0.6	0.2	1.2	12.2	1.2	24.7
July	Perimeter	47	1.1	0.1	2.4	18.2	4.0	29.5
	Off-Site	12	1.3	0.2	2.2	13.1	0.3	25.3
August	Perimeter	44	0.7	0.1	1.2	16.5	8.9	31.7
	Off-Site	14	0.9	0.2	1.6	16.8	2.2	30.5
September	Perimeter	58	0.9	0.4	2.1	21.2	9.6	42.3
	Off-Site	16	1.1	0.1	3.2	16.5	1.6	32.9
October	Perimeter	47	0.8	0.3	1.6	18.7	8.4	29.8
	Off-Site	14	1.1	0.1	2.5	19.1	6.0	34.4
November	Perimeter	48	1.6	0.7	3.4	29.6	16.5	50.9
	Off-Site	12	1.7	0.5	2.5	29.7	13.4	43.6
December	Perimeter	46	1.0	0.3	2.2	22.8	13.9	39.4
	Off-Site	13	1.6	0.4	3.1	27.3	8.9	45.4
Annual	Perimeter	571	0.9 ± 0.2	0.1	3.4	19.4 ± 2.8	4.0	50.9
summary	Off-Site	181	$1.1 \pm~0.2$	0.1	3.5	18.7 ± 4.1	0.3	57.6

TABLE 4.2

Gamma-Ray Activity in Air Filter Samples, 1999
(concentrations in fCi/m³)

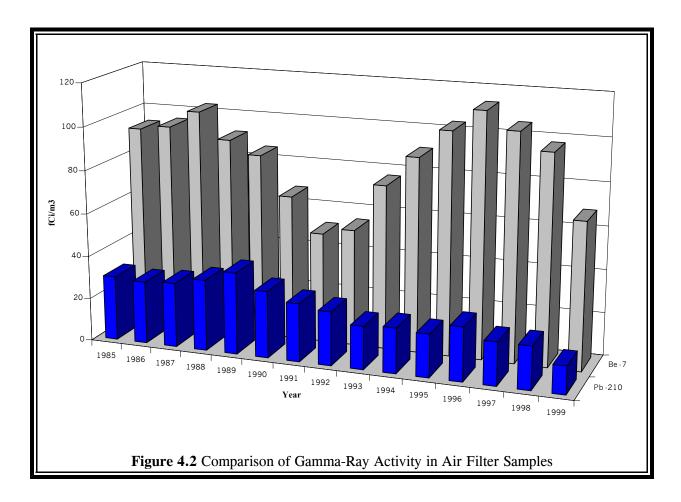
(concentrations in Tel/in)									
Month	Location	Beryllium-7	Lead-210						
January	Perimeter	69	21						
January	Off-Site	70	23						
	OII-Site	70	23						
February	Perimeter	89	17						
,	Off-Site	73	12						
March	Perimeter	110	15						
	Off-Site	80	11						
April	Perimeter	111	12						
	Off-Site	81	8						
May	Perimeter	107	11						
May	Off-Site	75	6						
	OII-Site	75	O						
June	Perimeter	95	13						
	Off-Site	66	8						
July	Perimeter	89	13						
	Off-Site	68	7						
August	Perimeter	88	14						
	Off-Site	69	11						
September	Perimeter	87	17						
Бертенност	Off-Site	56	10						
	OII BILL	30	10						
October	Perimeter	62	17						
	Off-Site	55	14						
November	Perimeter	62	26						
	Off-Site	64	23						
D 1	D : .	4.6	21						
December	Perimeter	46	21						
	Off-Site	56	21						
Annual	Perimeter	85 ± 13	16 ± 2						
Summary	Off-Site	68 ± 5	13 ± 3						
_ 		00 – 0	10 _ 0						
Dose(mrem)	Perimeter	(0.00021)	(1.87)						
	Off-Site	(0.00017)	(1.48)						



attributed to the Chernobyl incident are subtracted from the annual beta average of 40 fCi/m³, the net would be 27 fCi/m³, very similar to the averages of the other years. Figure 4.2 presents the annual average concentrations of the two major gamma-ray–emitting radionuclides in air. The annual average beryllium-7 concentrations have decreased regularly since 1987, reached a minimum in 1991, increased until 1996, and have now started to decrease. The changes in the beryllium-7 air concentrations have been observed worldwide by the DOE Environmental Laboratory's Surface Air Sampling Program and are attributed to changes in solar activity. ¹⁵

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Figure 1.1) and off the site in Downers Grove (Figure 1.2). Collections were made on polystyrene filters. The total air volume filtered for the monthly samples was approximately 20,000 m³ (700,000 ft³). Samples were ignited at 600°C (1,100°F) to remove organic matter and were prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids.

Plutonium and thorium were separated on an ion-exchange column, and the uranium was extracted from the column effluent. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242, thorium-229, and uranium-236 tracers prior to ignition. Because spectrometry cannot distinguish between



plutonium-239 and plutonium-240, when plutonium-239 is mentioned in this report, the alpha activity due to the plutonium-240 isotope is also included. The results are given in Table 4.3.

The strontium-90 concentrations have decreased over the past several years; consequently, during 1999, all of the results were less than the detection limit of 10 aCi/m³, except for two results in December. Strontium-89 was not observed above the detection limit of 100 aCi/m³. The plutonium-239 concentrations at all locations were similar to those of the last few years. The thorium and uranium concentrations were in the same range as in the past and are considered to be of natural origin. The amounts of thorium and uranium in a sample were proportional to the mass of inorganic material collected on the filter paper. The presence of most of these airborne elements can be attributed to the resuspension of soil.

The major airborne effluents released at ANL-E during 1999 are listed by location in Table 4.4; Figure 4.3 shows the annual releases of the major sources since 1985. The radon-220 releases from Building 200, due to radioactive contamination from the "proof-of-breeding" program, have been greatly reduced. The hydrogen-3 emitted from Building 212 is from hydrogen-3 recovery studies, while short-lived activation products are emitted from the IPNS and APS. In addition to the radionuclides listed in Table 4.4, several other fission products also were released in millicurie or

TABLE 4.3

Strontium, Thorium, Uranium, and Plutonium Concentrations in Air Filter Samples, 1999
(Concentrations in aCi/m³)

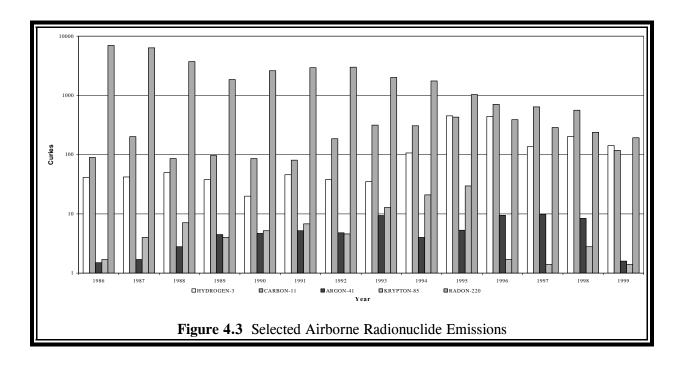
Month	Location ^a	Strontium-90	Thorium-228	Thorium-230	Thorium-232	Uranium-234	Uranium-238	Plutonium-239
January	7I	< 10	52 ± 6	39 ± 5	39 ± 5	47 ± 5	48 ± 5	2.1 ± 1.1
-	12N	< 10	23 ± 3	27 ± 3	19 ± 3	32 ± 3	32 ± 3	0.8 ± 0.5
	Off-Site	< 10	4 ± 1	5 ± 1	3 ± 1	8 ± 2	7 ± 2	0.2 ± 0.2
February	7I	< 10	7 ± 2	5 ± 1	4 ± 1	7 ± 2	8 ± 2	0.2 ± 0.4
	12N	< 10	13 ± 3	14 ± 3	11 ± 2	18 ± 4	19 ± 4	2.2 ± 1.2
	Off-Site	< 10	2 ± 1	2 ± 1	1 ± 1	5 ± 2	4 ± 1	< 0.1
March	7I	< 10	5 ± 2	5 ± 1	3 ± 1	8 ± 2	7 ± 2	0.3 ± 0.2
	12N	< 10	11 ± 2	12 ± 3	8 ± 2	17 ± 2	17 ± 2	0.4 ± 0.2
	Off-Site	< 10	6 ± 2	7 ± 2	4 ± 2	13 ± 4	8 ± 3	0.7 ± 0.6
April	7I	< 10	7 ± 2	7 ± 2	5 ± 1	8 ± 2	8 ± 2	0.3 ± 0.3
	12N	< 10	6 ± 1	6 ± 1	4 ± 1	7 ± 1	7 ± 1	0.3 ± 0.2
	Off-Site	< 10	2 ± 1	2 ± 1	1 ± 1	2 ± 1	2 ± 1	2.3 ± 0.7
May	7I	< 10	7 ± 2	7 ± 2	5 ± 1	9 ± 1	12 ± 1	0.3 ± 0.3
	12N	< 10	9 ± 2	9 ± 2	6 ± 1	11 ± 1	10 ± 1	-
	Off-Site	< 10	4 ± 2	5 ± 1	2 ± 1	7 ± 1	5 ± 1	0.5 ± 0.4
June	7I	< 10	3 ± 1	6 ± 2	3 ± 1	4 ± 1	4 ± 1	0.4 ± 0.2
	12N	< 10	7 ± 2	8 ± 2	6 ± 1	8 ± 1	8 ± 1	0.2 ± 0.1
	Off-Site	< 10	5 ± 2	5 ± 1	3 ± 1	6 ± 1	7 ± 1	0.2 ± 0.2
July	7I	< 10	3 ± 1	3 ± 1	1 ± 1	3 ± 1	4 ± 1	0.2 ± 0.2
	12N	< 10	8 ± 2	8 ± 2	6 ± 1	9 ± 1	10 ± 1	0.1 ± 0.1
	Off-Site	< 10	6 ± 2	5 ± 1	4 ± 1	6 ± 1	5 ± 1	0.3 ± 0.2
August	7I	< 10	5 ± 2	4 ± 1	3 ± 1	4 ± 1	5 ± 1	0.2 ± 0.2
	12N	< 10	5 ± 1	6 ± 1	4 ± 1	6 ± 1	5 ± 1	< 0.1
	Off-Site	< 10	3 ± 1	2 ± 1	1 ± 1	3 ± 1	3 ± 1	0.3 ± 0.2
September	7I	< 10	5 ± 2	4 ± 1	3 ± 1	5 ± 2	5 ± 1	< 0.1
	12N	< 10	7 ± 2	8 ± 2	5 ± 1	8 ± 1	8 ± 1	0.2 ± 0.2
	Off-Site	< 10	7 ± 2	8 ± 2	6 ± 1	7 ± 2	8 ± 2	0.3 ± 0.3
October	7I	< 10	3 ± 1	4 ± 1	2 ± 1	4 ± 1	4 ± 1	0.3 ± 0.3
	12N	< 10	8 ± 2	9 ± 2	7 ± 2	11 ± 2	10 ± 2	0.1 ± 0.1
	Off-Site	< 10	5 ± 2	5 ± 2	3 ± 1	5 ± 1	5 ± 1	0.2 ± 0.2
November	7I	< 10	2 ± 1	4 ± 1	2 ± 1	3 ± 1	4 ± 1	0.3 ± 0.3
	12N	< 10	9 ± 2	11 ± 2	8 ± 2	9 ± 1	10 ± 1	0.1 ± 0.2
	Off-Site	< 10	3 ± 1	3 ± 1	2 ± 1	3 ± 1	4 ± 1	0.3 ± 0.3
December	7I	16 ± 5	17 ± 7	17 ± 5	15 ± 5	16 ± 5	16 ± 5	1.7 ± 0.9
	12N	70 ± 16	14 ± 3	15 ± 2	10 ± 2	14 ± 2	15 ± 2	0.3 ± 0.3
	Off-Site	< 10	3 ± 1	5 ± 1	3 ± 1	5 ± 2	6 ± 2	0.4 ± 0.2
Annual	7I	< 10	10 ± 31	9 ± 22	7 ± 23	10 ± 27	10 ± 27	0.5 ± 1.5
Summary	12N	< 10	10 ± 11	11 ± 13	8 ± 9	12 ± 16	12 ± 16	0.4 ± 1.4
	Off-Site	< 10	4 ± 3	4 ± 4	3 ± 3	6 ± 6	5 ± 4	0.5 ± 1.3
Dose	7I	< (0.00011)	(0.0238)	(0.0172)	(0.071)	(0.00049)	(0.00051)	(0.0013)
(mrem)	12N	< (0.00011)	(0.0245)	(0.0221)	(0.078)	(0.00062)	(0.00062)	(0.0011)
	Off-Site	< (0.00011)	(0.0099)	(0.0089)	(0.027)	(0.00029)	(0.00026)	(0.0012)

^a Perimeter locations are given in terms of the grid coordinates in Figure 1.1

TABLE 4.4

Summary of Monitored Airborne Radioactive Emissions from ANL-E Facilities, 1999

Building	Nuclide	Half-Life	Amount Released (Ci)	Amount Released (Bq)
200	Radon-220	56 s	193.0	7.1×10^{12}
205	Hydrogen-3 (tritiated water [HTO])	12.3 yr	0.53	1.9×10^{10}
212 (Alpha	Hydrogen-3 (HTO)	12.3 yr	10.9	4.0×10^{11}
Gamma Hot Cell Facility)	Hydrogen-3 (tritiated hydrogen gas [HT])	12.3 yr	131.4	4.8×10^{12}
	Krypton-85	10.7 yr	1.35	5.0×10^{10}
	Radon-220	56 s	0.14	5.2×10^{9}
350 (NBL)	Uranium-234	$2.4 \times 10^5 \text{ yr}$	3.2×10^{-5}	1.2×10^{1}
	Uranium-238	$4.5 \times 10^9 \text{ yr}$	3.2×10^{-5}	1.2×10^{1}
	Plutonium-238	87.7 yr	1.5×10^{-9}	5.5×10^{0}
	Plutonium-239	$2.4 \times 10^4 \text{ yr}$	5.8×10^{-9}	2.1×10^{1}
	Plutonium-240	$6.6 \times 10^4 \text{ yr}$	8.5×10^{-9}	3.1×10^{1}
	Plutonium-241	14.4 yr	2.2×10^{-7}	8.1×10^{3}
	Plutonium-242	$3.76 \times 10^{5} \text{ yr}$	1.8×10^{-9}	6.6×10^{0}
	Plutonium-244	$8.0 \times 10^7 \text{ yr}$	3.3×10^{-13}	1.2×10^{-3}
375 (IPNS)	Carbon-11	20 m	118.1	4.3×10^{12}
	Argon-41	1.8 h	1.6	5.9×10^{10}
411/415 (APS)	Carbon-11	20 m	0.07	2.6×10^{9}
,	Nitrogen-13	10 m	3.22	1.2×10^{11}
	Oxygen-15	122 s	0.35	1.3×10^{10}



smaller amounts. The quantities listed in Table 4.4 were measured by on-line stack monitors in the exhaust systems of the buildings, except those for Building 350.

4.3. Surface Water

All water samples collected in the monitoring program were acidified to $0.1\underline{N}$ with nitric acid and filtered immediately after collection. Total nonvolatile alpha and beta activities were determined by counting the residue remaining after evaporation of the water and then applying counting efficiency corrections determined for plutonium-239 (for alpha activity) and thallium-204 (for beta activity) to obtain disintegration rates. Hydrogen-3 was measured from a separate aliquot; this activity does not appear in the results for total nonvolatile beta activity. Analyses for the radionuclides were performed by specific radiochemical separations followed by appropriate counting. One-liter aliquots were used for all analyses except for hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by liquid scintillation counting of 9 mL (0.03 oz) of a distilled sample in a nonhazardous cocktail. Analyses for transuranium nuclides were performed on 10-L (3-gal) samples with chemical separation methods followed by alpha spectrometry. Plutonium-236 was used to determine the yields of plutonium and neptunium, which were separated from the sample together. A group separation of a fraction containing the transplutonium elements was monitored for recovery with an americium-243 tracer. Isotopic uranium concentrations were determined by alpha spectrometry by using uranium-236 as an isotopic tracer.

Liquid wastewater from buildings or facilities that use or process radioactive materials are collected in retention tanks. When a tank is full, it is sampled and analyzed for alpha and beta

radioactivity. If the radioactivity exceeds the release limits, the tank is processed by evaporation and the residue is disposed of as solid LLW. If the radioactivity is below the release limits, the wastewater is conveyed to the laboratory WTP in dedicated pipes to waste storage tanks. The release limits are based on the DCGs for plutonium-239 (0.03 pCi/mL) for alpha activity, and for strontium-90 (1.0 pCi/mL) for beta activity. These radionuclides were selected because of their potential for release and their conservative allowable limits in the environment. The effluent monitoring program documents that no liquid releases above the DCGs have occurred and reinforces demonstration of compliance with the use of BAT as required by DOE Order 5400.5.¹¹

Another component of the radiological effluent monitoring program, which was instituted in 1999, is the radiological analysis of the main water treatment plant discharge (Outfall 001). Metals have been analyzed at this location for a number of years (see Table 5.10). The same constituents that are determined in Sawmill Creek are also analyzed at this location. Samples are collected daily, and equal portions are combined for each week and analyzed to obtain an average weekly concentration. Table 4.5 gives the results for 1999. The results show that the radionuclides hydrogen-3 and strontium-90 detected in the effluent water can be attributed to ANL-E operations. The concentrations are very low and a small fraction of the DOE limits; these findings reinforce ANL-E compliance with DOE Order 5400.5 for use of BAT for releases of liquid effluents.

To estimate the total annual quantity of each radionuclide released to the environment, the product of the annual average concentration and the annual volume of water discharged $(1.08 \times 10^9 \text{ L})$ is computed. These results are given in Table 4.6, along with similar data from concentrations measured in Sawmill Creek.

ANL-E wastewater is discharged into Sawmill Creek at Location 7M (Figure 1.1). The creek runs through the ANL-E grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (1,600 ft) downstream from the ANL-E wastewater outfall. Sawmill Creek was sampled upstream from the ANL-E site and downstream from the wastewater outfall to determine whether radioactivity was added to the stream by ANL-E wastewater or surface drainage. The sampling locations are shown in Figure 1.1. Below the wastewater outfall, daily samples were collected. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Samples were collected upstream of the site once a month and were analyzed for the same radionuclides measured in the below-outfall samples.

Table 4.7 gives the annual summaries of the results obtained for Sawmill Creek. Comparison of the results and 95% confidence levels of the averages for the two sampling locations shows that the following radionuclides found in the creek water can be attributed to ANL-E operations: hydrogen-3, strontium-90, plutonium-239, and americium-241; and occasionally neptunium-237, plutonium-238, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to ANL-E was 65% for hydrogen-3, 92% for

TABLE 4.5

Radionuclides in Effluents from the ANL-E Wastewater Treatment Plant, 1999

		Concentrations in pCi/L			Dose (mrem)				
Activity	No. of Samples	Avg.	Min.	Max.	Avg.	Min.	Max.		
Alpha	52	0.5 ± 0.7	< 0.1	1.4	_a	-	-		
Beta	52	15 ± 5	9	21	-	-	-		
Hydrogen-3	52	626 ± 3652	< 100	12780	0.0287	< 0.0046	0.5866		
Strontium-90	52	0.77 ± 1.10	0.45	3.76	0.073	0.043	0.357		
Cesium-137	52	< 1.0	< 1.0	< 1.0	< 0.04	< 0.04	< 0.04		
Uranium-234	52	0.261 ± 0.358	0.083	0.714	0.050	0.016	0.136		
Uranium-238	52	0.233 ± 0.326	0.067	0.712	0.039	0.011	0.120		
Neptunium-237	52	< 0.0010	< 0.0010	0.0033	< 0.0028	< 0.0028	0.0094		
Plutonium-238	52	< 0.0010	< 0.0010	0.0048	< 0.0028	< 0.0028	0.0134		
Plutonium-239	52	< 0.0010	< 0.0010	0.0068	< 0.0031	< 0.0031	0.0214		
Americium-241	52	< 0.0010	< 0.0010	0.0029	< 0.0033	< 0.0033	0.0097		
Curium-242 and/or Californium - 252	52	< 0.0010	< 0.0010	< 0.0010	< 0.0007	< 0.0007	< 0.0007		
Curium-244 and/or Californium - 249	52	< 0.0010	< 0.0010	0.0010	< 0.0034	< 0.0034	0.0034		

^a A hyphen indicates no CEDEs for alpha and beta.

strontium-90, 24% for plutonium-239, and 67% for americium-241. The concentrations of all these nuclides are low and at a small fraction of DOE limits.

At location 7M, below the ANL-E outfall, the annual average concentrations of most measured radionuclides were similar to recent annual averages. All the annual averages were well below the applicable standards. The annual total radioactive effluent discharged to the creek in ANL-E wastewater can be estimated from the average net concentrations and the volume of water carried by the creek. These totals are presented in Table 4.6. Comparison of the total quantity released, as calculated from measurements at the Outfall 001 and in Sawmill Creek, indicate

significant differences. The differences in volume between the two locations is about 10, while the concentrations differ by a factor of 2. It is speculated that the measurement of the Sawmill Creek flow has the greatest uncertainty.

On the basis of the results of the Storm Water Characterization Study (see Section 2.2.2), two perimeter surface water locations were identified that contained measurable levels of radionuclides. They were south of the 319 Area, Location 7J, and south

TABLE 4.6

Total Radioactivity Released to Sawmill Creek, 1999

Radionuclide	Outfall (Ci)	Creek (Ci)
Hydrogen-3	0.68	4.44
Strontium-90	0.0006	0.0037
Plutonium-239	< 0.0001	< 0.0001
Americium-241	< 0.0001	< 0.0001
Total	0.68	4.44

of the 800 Area Landfill, Location 11D (see Figure 1.1). Samples were scheduled to be collected quarterly and analyzed for hydrogen-3 and strontium-90 and by gamma-ray spectrometry. The results are presented in Table 4.8.

The source of the radionuclides at Location 7J appears to be leachate from the 319 Area Landfill. A subsurface barrier wall and leachate collection system were constructed south of the 319 Landfill in November 1995 and became operational in 1996. Since the construction and operation of the leachate collection system, radionuclide concentrations in surface water at Location 7J have decreased substantially. The hydrogen-3 at Location 11D is probably also from the leachate; the decrease in the concentration from earlier years is due to the completion of the clay cap on the 800 Area Landfill in the fall of 1993.

Because Sawmill Creek empties into the Des Plaines River, data on the radioactivity in this river is important in assessing the contribution of ANL-E wastewater to environmental radioactivity. The Des Plaines River was sampled twice a month below and once a month above the mouth of Sawmill Creek to determine whether the radioactivity in the creek had any effect on the radioactivity in the river.

Table 4.9 gives the annual summaries of the results obtained for these two locations. The average nonvolatile alpha, beta, and uranium concentrations in the river were very similar to past averages and remained in the normal range. Results were quite similar above and below the creek for all radionuclides, because the activity in Sawmill Creek was reduced by dilution to the point that it was not detectable in the Des Plaines River.

TABLE 4.7

Radionuclides in Sawmill Creek Water, 1999

			Concentrations in pCi/L				Dose (mrem)	
Activity	Location ^a	No. of Samples	Avg.	Min.	Max.	Avg.	Min.	Max.
Alpha	16K	12	1.1 ± 1.0	0.3	2.2	_b	-	-
(Nonvolatile)	7M	49	1.2 ± 1.1	0.2	2.7	-	-	-
Beta	16K	12	7 ± 5	4	12	-	-	-
(Nonvolatile)	7M	49	12 ± 6	6	20	-	-	-
Hydrogen-3	16K	12	< 100	< 100	109	< 0.0046	< 0.0046	0.0050
	7M	49	418 ± 3608	< 100	12700	0.0192	< 0.0046	0.5829
Strontium-90	16K	12	< 0.25	< 0.25	0.32	< 0.024	< 0.024	0.031
	7 M	49	0.56 ± 0.93	< 0.25	3.25	0.053	< 0.024	0.308
Cesium-137	16K	12	< 1.0	< 1.0	< 1.0	< 0.04	< 0.04	< 0.04
	7M	49	< 1.0	< 1.0	< 1.0	< 0.04	< 0.04	< 0.04
Uranium-234	16K	12	0.574 ± 0.573	0.275	0.976	0.109	0.052	0.185
	7M	49	0.450 ± 0.517	0.114	1.044	0.086	0.022	0.198
Uranium-238	16K	12	0.537 ± 0.561	0.241	1.004	0.090	0.040	0.169
	7M	49	0.401 ± 0.481	0.087	0.928	0.067	0.015	0.156
Neptunium-237	16K	12	< 0.0010	< 0.0010	< 0.0010	< 0.0028	< 0.0028	< 0.0028
-	7M	49	< 0.0010	< 0.0010	0.0011	< 0.0028	< 0.0028	0.0032
Plutonium-238	16K	12	< 0.0010	< 0.0010	0.0019	< 0.0028	< 0.0028	0.0051
	7M	49	< 0.0010	< 0.0010	0.0019	< 0.0028	< 0.0028	0.0053
Plutonium-239	16K	12	< 0.0010	< 0.0010	< 0.0010	< 0.0031	< 0.0031	< 0.0031
	7M	49	< 0.0010	< 0.0010	0.0019	< 0.0031	< 0.0031	0.0059
Americium-241	16K	12	< 0.0010	< 0.0010	< 0.0010	< 0.0033	< 0.0033	< 0.0033
	7M	49	0.0014 ± 0.0018	< 0.0010	0.0046	0.0045	< 0.0033	0.0150
Curium-242 and/or	16K	12	< 0.0010	< 0.0010	< 0.0010	< 0.0007	< 0.0007	< 0.0007
Californium-252	7M	49	< 0.0010	< 0.0010	0.0011	< 0.0007	< 0.0007	0.0007
Curium-244 and/or	16K	12	< 0.0010	< 0.0010	< 0.0010	< 0.0034	< 0.0034	< 0.0034
Californium-249	7M	49	< 0.0010	< 0.0010	0.0011	< 0.0034	< 0.0034	0.0037

^a Location 16K is upstream from the ANL-E site, and location 7M is downstream from the ANL-E wastewater outfall.

^b A hyphen indicates no CEDEs for alpha and beta.

TABLE 4.8

Radionuclides in Storm Water Outfalls, 1999
(concentrations in pCi/L)

Date Collected	Location 7J Hydrogen-3	Location 7J Strontium-90	Location 7J Cesium-137	Location 11D Hydrogen-3
January 27	<100	0.63	<1	373
April 16	<100	0.63	<1	646
August 25	278	0.86	<1	Dry
October	Dry	Dry	Dry	Dry

4.4. Bottom Sediment

The radioactive content of bottom sediment was measured in Sawmill Creek. A grab sample technique was used to obtain bottom sediments. After drying, grinding, and mixing, portions of each of the bottom sediment samples were analyzed by the same methods described in Section 4.2 for air filter residues. The plutonium and americium were separated from the same 10-g (0.35-oz) aliquot of soil. Results are given in terms of the oven-dried (110EC [230EF]) weight.

A set of sediment samples was collected on October 27, 1999, from the Sawmill Creek bed, above, at the outfall, and at several locations below the point at which ANL-E discharges its treated wastewater (location 7M in Figure 1.1). The results, as listed in Table 4.10, show that the concentrations in the samples collected above the 7M outfall are similar to those of the off-site samples collected in past years. The plutonium, americium, and cesium-137 concentrations are elevated below the outfall, which indicates that their origin is in ANL-E wastewater. Plutonium results varied widely among locations and were strongly dependent on the retentiveness of the bottom material. The changes in concentrations of these nuclides with time and location indicate the dynamic nature of the sediment material in this area.

4.5. External Penetrating Radiation

Levels of external penetrating radiation at and in the vicinity of the ANL-E site were measured with aluminum oxide TLD chips provided and read by a commercial vendor. Each measurement reported represents the average of two chips exposed in the same packet. Dosimeters were exposed at 17 locations at the site boundary and on the site. Readings were also taken at five off-site locations (Figure 1.2) for comparative purposes. Three locations were added to the network in 1999 to monitor radioactive waste management activities. They are east of Building 306

TABLE 4.9

Radionuclides in Des Plaines River Water, 1999

		No. of Samples	Conce	Concentrations in pCi/L			Dose (mrem)		
Activity	Location ^a		Avg.	Min.	Max.	Avg.	Min.	Max.	
Alpha	A	12	0.9 ± 0.7	0.5	1.5	_b	-	-	
(Nonvolatile)	В	24	1.1 ± 1.5	0.3	4.2	-	-	-	
Beta	A	12	16 ± 13	8	25	-	-	-	
(Nonvolatile)	В	24	16 ± 11	7	25	-	-	-	
Hydrogen-3	A	12	< 100	< 100	107	< 0.0046	< 0.0046	0.0049	
	В	24	< 100	< 100	107	< 0.0046	< 0.0046	0.0049	
Strontium-90	A	12	0.27 ± 0.14	< 0.25	0.37	0.025	< 0.024	0.036	
	В	24	0.28 ± 0.13	< 0.25	0.39	0.027	< 0.024	0.037	
Uranium-234	A	12	0.496 ± 0.434	0.271	0.893	0.094	0.051	0.170	
	В	24	0.476 ± 0.437	0.122	0.824	0.090	0.023	0.157	
Uranium-238	A	12	0.405 ± 0.387	0.227	0.735	0.068	0.038	0.124	
	В	24	0.396 ± 0.404	0.089	0.748	0.066	0.015	0.126	
Neptunium-237	A	12	< 0.0010	< 0.0010	0.0033	< 0.0028	< 0.0028	0.0094	
·	В	12	< 0.0010	< 0.0010	< 0.0010	< 0.0028	< 0.0028	< 0.0028	
Plutonium-238	A	12	< 0.0010	< 0.0010	0.0033	< 0.0028	< 0.0028	0.0090	
	В	12	< 0.0010	< 0.0010	< 0.0010	< 0.0028	< 0.0028	< 0.0028	
Plutonium-239	A	12	< 0.0010	< 0.0010	< 0.0010	< 0.0031	< 0.0031	< 0.0031	
	В	11	< 0.0010	< 0.0010	< 0.0010	< 0.0031	< 0.0031	< 0.0031	
Americium-241	A	12	< 0.0010	< 0.0010	0.0034	< 0.0033	< 0.0033	0.0110	
	В	12	< 0.0010	< 0.0010	0.0046	< 0.0033	< 0.0033	0.0151	
Curium-242 and/or	A	12	< 0.0010	< 0.0010	0.0013	< 0.0007	< 0.0007	0.0009	
Californium-252	В	12	< 0.0010	< 0.0010	< 0.0010	< 0.0007	< 0.0007	< 0.0007	
Curium-244 and/or	A	12	< 0.0010	< 0.0010	< 0.0010	< 0.0034	< 0.0034	< 0.0034	
Californium-249	В	12	< 0.0010	< 0.0010	< 0.0010	< 0.0034	< 0.0034	< 0.0034	

^a Location A, near Willow Springs, is upstream; location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 1.2

^b A hyphen indicates no CEDEs for alpha and beta.

TABLE 4.10

Radionuclides in Bottom Sediment, 1999^a

			Concentrations (pCi/g)		Concentrations (fCi/g)			
Location	Potassium-40	Cesium-137	Radium-226	Thorium-228	Thorium-232	Plutonium-238	Plutonium-239	Americium-241
Sawmill Creek 25 m above Outfall	8.66 ± 0.48	0.02 ± 0.02	0.50 ± 0.05	0.36 ± 0.04	0.24 ± 0.07	< 0.1	1.5 ± 0.7	0.8 ± 0.3
Sawmill Creek at Outfall	5.23 ± 0.40	0.37 ± 0.03	0.68 ± 0.06	0.55 ± 0.04	0.44 ± 0.08	47.2 ± 3.8	361.8 ± 20.5	117.5 ± 6.4
Sawmill Creek 50 m below Outfall	9.50 ± 0.49	0.10 ± 0.02	0.54 ± 0.06	0.44 ± 0.04	0.28 ± 0.07	0.4 ± 0.3	9.4 ± 1.6	3.4 ± 0.8
Sawmill Creek 100 m below Outfall	< 0.01	0.10 ± 0.02	0.49 ± 0.05	0.31 ± 0.03	0.26 ± 0.07	0.4 ± 0.3	6.1 ± 1.3	2.0 ± 0.5
Sawmill Creek at Des Plaines River	12.69 ± 0.56	0.24 ± 0.03	0.63 ± 0.06	0.58 ± 0.04	0.43 ± 0.08	1.4 ± 0.6	21.3 ± 2.5	5.7 ± 1.0

^a All samples were collected on October 27, 1999.

(Location 9/10 I), south of Building 331 (Location 9 H/I), and next to the 398A radioactive waste storage area (Location 9J).

The results are summarized in Tables 4.11 and 4.12, and the site boundary and on-site readings are shown in Figure 4.4. Measurements were taken during the four successive exposure periods shown in the tables, and the results were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times. The uncertainty of the averages given in the tables is the 95% confidence limit calculated from the standard deviation of the average.

The off-site results averaged 80 ± 4 mrem/yr and were identical to last year's off-site average of 80 ± 3 mrem/yr. ¹⁶ To compare boundary results for individual sampling periods, the standard deviation of the 19 individual off-site results is useful. This value is 8 mrem/yr; thus, individual results in the range of 80 ± 16 mrem/yr may be considered to be the average natural background with a 95% probability.

The site boundary location at 7I had dose rates consistently above the average background. This was the result of radiation from ANL-E's 317 Area in the northern half of grid 7I. Waste is packaged and temporarily stored in this area before removal for permanent disposal off site. In 1999, the dose at this perimeter fence location was 105 ± 21 mrem/yr. Approximately 300 m (960 ft) south of the fence in grid 6I, the measured dose dropped to 88 ± 11 mrem/yr, which is within the normal background range.

TABLE 4.11

Environmental Penetrating Radiation at Off-Site Locations, 1999

	Dose Rate (mrem/year) Period of Measurement										
Location	Jan. 7 – April 1	April 1 – July 6	July 6 – Oct. 6	Oct. 6 – Jan. 6	Average						
Lemont	77	89	90	64	80 ± 12						
Oak Brook	80	89	100	77	87 ± 10						
Orland Park	76	80	64	_a	73 ± 9						
Woodridge	90	91	76	77	84 ± 8						
Willow Springs	79	70	78	65	73 ± 7						
Average	80 ± 5	84 ± 8	82 ± 12	71 ± 7	80 ± 4						

^a Sample lost.

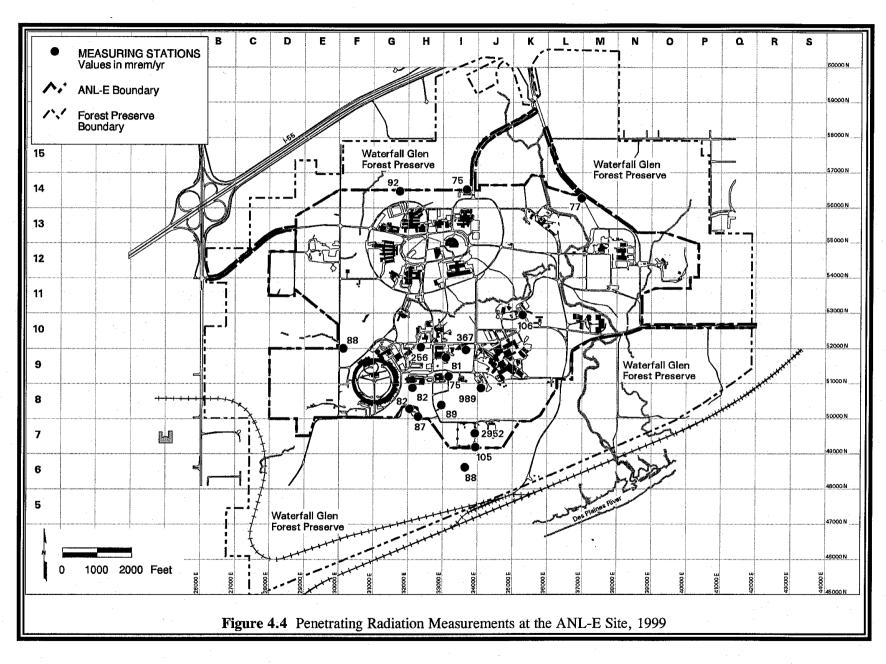
TABLE 4.12Environmental Penetrating Radiation at ANL-E, 1999

Dose Rate (mrem/year) Period of Measurement

Location ^a	Jan. 7 – April 1	April 1 – July 6	July 6 – Oct. 6	Oct. 6 – Jan. 6	Average
14G - Boundary	101	98	94	73	92 ± 12
14I - Boundary	89	54	79	78	75 ± 15
14L - Boundary	77	65	94	72	77 ± 12
6I - 200 m N of Quarry Road	98	89	93	73	88 ± 11
7I - Center, Waste Storage Area Facility 317	3,614	2,467	3,051	2,676	$2,952 \pm 493$
7I - Boundary	117	79	126	96	105 ± 21
8H - Boundary	96	97	85	70	87 ± 12
8H - 65 m S of Building 316	89	71	97	69	82 ± 13
8H - 200 m NW of Waste Storage Area (Heliport)	89	81	111	75	89 ± 15
8H - Boundary, Center, St. Patrick Cemetery	85	86	75	_b	82 ± 7
9H - 50 m SE of CP-5	129	232	540	121	256 ± 192
9 H/I - 50 m E of Building 331	176	68	720	504	367 ± 294
9/10 I - E of D306	75	75	77	96	81 ± 10
9/10 I - 65 m NE of Building 350, 230 m NE of Building 316	66	78	81	73	75 ± 6
9/10 IF - Boundary	106	61	103	81	88 ± 21
9J - 50 m W of 398A Area	772	699	859	1,625	989 ± 421
10/11 K - Facilities	89	186	84	66	106 ± 53

^a See Figure 1.1

^b This sample was lost.



In the past, an elevated on-site dose had been measured at Location 9H, next to the CP-5 reactor, where irradiated hardware from the CP-5 reactor was stored. During the past few years, considerable cleanup of the CP-5 reactor yard has occurred as part of the CP-5 reactor D&D project. The dose at Location 9H decreased from about 1,200 mrem/yr in 1989 to 256 mrem/yr in 1999. The cleanup of the yard was completed in 1994; the residual dose is from sources in the building, which is currently undergoing D&D, and the use of the yard to stage radioactive waste from the D&D, pending shipment off site.

The three new locations were added to monitor radioactive waste facilities and areas. Significant movement of radioactive waste took place, principally waste from the D&D of the CP-5 reactor and the relocation of radioactive waste from the 317 Area to the 398A Area. Current plans call for evacuating the 317 Area so that it can be remediated. Although some waste is repacked in Building 306 (Location 9/10 I), the dose from these operations could not be distinguished from normal background levels. The elevated dose levels in the 398A Area (Location 9J) are from waste relocated from the 317 Area, historic waste, and D&D waste temporarily stored pending shipment. The Building 331 yard (Location 9 H/I) is being used as a staging area to load trucks for shipment off site. A large number of radioactive waste shipments were made during the second half of 1999, as reflected by the elevated dose rates. Much of this waste came from the D&D of the CP-5 reactor, as indicated by the elevated doses in the second and third quarters when waste was stored in the CP-5 yard pending transfer to the Building 331 yard for shipment. The second quarter result of 186 mrem/yr at location 10/11 K appears to be an outlier.

4.6. Estimates of Potential Radiation Doses

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials and radiation leaving the site were calculated. Calculations were performed for three exposure pathways—airborne, water, and direct radiation from external sources.

4.6.1. Airborne Pathway

Guidance issued by DOE¹¹ stipulates that DOE facilities with airborne releases of radioactive materials are subject to 40 CFR Part 61, Subpart H,¹⁷ which requires the use of the EPA's CAP-88 code⁸ to calculate the dose for radionuclides released to the air and to demonstrate compliance with the regulation. The dose limit applicable for 1999 for the air pathway is a 10-mrem/yr effective dose equivalent. The CAP-88 computer code uses a modified Gaussian plume equation to estimate both horizontal and vertical dispersion of radionuclides released to the air from stacks or area sources. For 1999, doses were calculated for hydrogen-3, carbon-11, nitrogen-13, oxygen-15, argon-41, krypton-85, radon-220 plus daughters, and a number of actinide radionuclides.

The annual releases are those listed in Table 4.4; separate calculations were performed for each of the six release points. The wind speed and direction data shown in Figure 1.3 were used for these calculations. In the past, the wind stability classes had been determined by the temperature differences between the 10-m (33-ft) and 60-m (197-ft) levels. To improve the determination of stability levels, the categories were obtained from daytime measurements of solar radiation and nighttime measurements of the standard deviation of the horizontal wind speed. Doses were calculated for an area extending out to 80 km (50 mi) from ANL-E. The population distribution of the 16 compass segments and 10 distance increments given in Table 1.1 was used. The dose rate was calculated at the midpoint of each interval and integrated over the entire area to give the annual population cumulative dose.

Distances from the specific facilities that exhaust radiological airborne emissions (see Table 4.4) to the fence line (perimeter) and nearest resident were determined in the 16 compass segments. Calculations also were performed to evaluate the major airborne pathways — ingestion, inhalation, and immersion — both at the point of maximum perimeter exposure and to the maximally exposed resident. The perimeter and resident doses and the maximum doses are listed, respectively, for releases from Buildings 200 (Tables 4.13 and 4.14), Building 205 (Tables 4.15 and 4.16), Building 212 (Tables 4.17 and 4.18), Building 350 (Tables 4.19 and 4.20), Building 375 (Tables 4.21 and 4.22), and Building 411 (Tables 4.23 and 4.24). The doses given in these tables are the committed whole body effective dose equivalents.

A significant D&D program was completed for the M-Wing hot cells in Building 200, which constituted the source of the radon-220 emissions. Cleanup of the major source of the radon-220, cell M-1, was completed in 1995. This has resulted in a decrease of radon-220 emissions: 3,000 Ci in 1992; 2,023 Ci in 1993; 1,750 Ci in 1994; 1,033 Ci in 1995; 388 Ci in 1996; 286.6 Ci in 1997; 239.2 Ci in 1998; and 193.0 Ci in 1999. The present radon-220 emissions will be reduced because of the termination of the nuclear medical program that separates radium-224 from the thorium-228 parent and continued D&D of other cells. The highest perimeter dose was in the southwest direction with a maximum dose of 0.063 mrem/yr (location 7H in Figure 1.1). The major component of this dose is air immersion of carbon-11 (0.032 mrem/yr).

The full-time resident who would receive the largest annual dose (0.015 mrem/yr) is located approximately 0.8 km (0.5 mi) north-northwest of the site boundary. The major contributor to the whole body dose is the inhalation dose from lead-212 (0.009 mrem/yr). If radon-220 plus daughters were excluded from the calculation, as required by NESHAP,¹⁷ the maximally exposed resident would receive a dose of 0.0043 mrem/yr, primarily carbon-11 from the IPNS facility (Building 375).

TABLE 4.13

Radiological Airborne Releases from Building 200, 1999

	Distance to Distance to Nearest				
	Perimeter	Dose ^a	Resident	Dose ^a	
Direction	(m)	(mrem/yr)	(m)	(mrem/yr)	
N	500	4.1×10^{-2}	1,000	1.2×10^{-2}	
NNE	600	3.3×10^{-2}	1,100	1.0×10^{-2}	
NE	750	1.9×10^{-2}	2,600	2.0×10^{-3}	
ENE	1,700	4.0×10^{-3}	3,100	1.4×10^{-3}	
E	2,400	2.9×10^{-3}	3,500	1.5×10^{-3}	
ESE	2,200	2.4×10^{-3}	3,600	1.0×10^{-3}	
SE	2,100	2.0×10^{-3}	4,000	6.8×10^{-4}	
SSE	2,000	2.9×10^{-3}	4,000	9.1×10^{-4}	
S	1,500	2.4×10^{-3}	4,000	5.0×10^{-4}	
SSW	1,000	1.3×10^{-2}	2,500	2.7×10^{-3}	
SW	800	2.7×10^{-2}	2,200	5.6×10^{-3}	
WSW	1,100	8.5×10^{-3}	1,500	4.9×10^{-3}	
W	750	1.5×10^{-2}	1,500	5.0×10^{-3}	
WNW	800	9.8×10^{-3}	1,300	4.6×10^{-3}	
NW	600	1.5×10^{-2}	1,100	5.6×10^{-3}	
NNW	600	2.1×10^{-2}	800	1.2×10^{-2}	

^a Source term: radon-220 = 193.0 Ci (plus daughters).

TABLE 4.14

Maximum Perimeter and Individual Doses from Building 200 Air Emissions, 1999 (dose in mrem/yr)

Pathway	Perimeter (500 m N)	Individual (800 m NNW)
Ingestion	5.9×10^{-14}	2.2× 10 ⁻¹⁴
Inhalation	4.1×10^{-2}	1.2×10^{-2}
Air immersion	2.8×10^{-4}	7.5×10^{-5}
Ground surface	2.0×10^{-5}	7.4×10^{-6}
Total	4.1×10^{-2}	1.2×10^{-2}
Radionuclide		
Thallium-208	2.5×10^{-4}	6.3×10^{-5}
Bismuth-212	4.9×10^{-3}	1.7×10^{-3}
Lead-212	2.4×10^{-2}	8.7×10^{-3}
Radon-220	1.2×10^{-2}	1.7×10^{-3}
Total	4.1×10^{-2}	1.2×10^{-2}

TABLE 4.15

Radiological Airborne Releases from Building 205, 1999

Direction	Distance to Perimeter (m)	Dose ^a (mrem/yr)	Distance to Nearest Resident (m)	Dose ^a (mrem/yr)
N	850	3.3×10^{-5}	1,300	1.6×10^{-5}
NNE	1,000	2.6×10^{-5}	2,100	7.5×10^{-6}
NE	1,200	1.7×10^{-5}	2,700	4.5×10^{-6}
ENE	2,400	5.2×10^{-6}	3,000	3.6×10^{-6}
E	2,200	7.9×10^{-6}	2,400	3.9×10^{-6}
ESE	2,000	6.5×10^{-6}	3,500	2.6×10^{-6}
SE	1,800	6.1×10^{-6}	3,900	1.7×10^{-6}
SSE	1,500	1.1×10^{-5}	4,000	2.2×10^{-6}
S	1,300	7.3×10^{-6}	3,900	1.3×10^{-6}
SSW	1,100	2.6×10^{-5}	2,400	7.1×10^{-6}
SW	900	5.9×10^{-5}	2,100	1.8×10^{-5}
WSW	1,100	$1.9\times10^{\text{-5}}$	1,800	8.5×10^{-6}
W	1,300	1.3×10^{-5}	1,800	9.5×10^{-6}
WNW	1,100	$1.5\times10^{\text{-5}}$	1,700	7.6×10^{-6}
NW	1,100	$1.4\times10^{\text{-5}}$	1,500	8.4×10^{-6}
NNW	900	2.2×10^{-5}	1,500	9.3×10^{-6}

^a Source term: hydrogen-3 = 0.53 Ci.

TABLE 4.16

Maximum Perimeter and Individual Doses from Building 205 Air Emissions, 1999
(dose in mrem/yr)

Pathway	Perimeter (900 m SW)	Individual (2,100 m SW)
Ingostion	1.4×10^{-5}	4.4×10^{-6}
Ingestion	1.4×10	4.4×10
Inhalation	4.5×10^{-5}	1.4×10^{-5}
Air immersion	_a	-
Ground surface	-	-
Total	5.9×10^{-5}	1.8×10^{-5}
Radionuclide		
Hydrogen-3	5.9×10^{-5}	1.8×10^{-5}

^a A hyphen indicates no exposure by this pathway.

TABLE 4.17Radiological Airborne Releases from Building 212, 1999

	Distance to Perimeter	Dose ^a	Distance to Nearest Resident	Dose ^a
Direction	(m)	(mrem/yr)	(m)	(mrem/yr)
Direction	(III)	(IIIICIII/y1)	(III)	(IIIICIII/yI)
N	800	5.7×10^{-3}	2,000	1.4×10^{-3}
NNE	1,000	4.2×10^{-3}	2,500	9.9×10^{-4}
NE	1,300	2.5×10^{-3}	2,000	1.3×10^{-3}
ENE	1,500	1.9×10^{-3}	2,500	8.6×10^{-4}
E	1,600	2.2×10^{-3}	2,800	9.3×10^{-4}
ESE	1,200	2.6×10^{-3}	2,500	7.9×10^{-4}
SE	1,400	1.5×10^{-3}	3,500	3.6×10^{-4}
SSE	1,400	2.1×10^{-3}	4,500	3.4×10^{-4}
S	1,500	9.4×10^{-4}	5,000	1.6×10^{-4}
SSW	1,600	2.3×10^{-3}	5,000	4.1×10^{-4}
SW	1,400	4.2×10^{-3}	2,400	2.0×10^{-3}
WSW	1,300	2.4×10^{-3}	2,300	9.8×10^{-4}
W	1,700	1.6×10^{-3}	2,200	1.1×10^{-3}
WNW	1,500	1.4×10^{-3}	2,000	9.3×10^{-4}
NW	1,300	1.6×10^{-3}	2,000	8.6×10^{-4}
NNW	1,000	2.9×10^{-3}	2,000	9.9×10^{-4}

 a Source terms: hydrogen-3 (HT) = 131.4 Ci

hydrogen-3 (HTO) = 10.9 Ci krypton-85 = 1.35 Ci radon-220 = 0.14 Ci.

TABLE 4.18

Maximum Perimeter and Individual Doses from Building 212 Air Emissions, 1999 (dose in mrem/yr)

Pathway	Perimeter (800 m N)	Individual (2,400 m SW)
Ingestion	1.3×10^{-3}	4.8×10^{-4}
Inhalation	4.3×10^{-3}	1.5×10^{-3}
Air immersion	4.5×10^{-7}	1.6×10^{-7}
Ground surface	3.8×10^{-8}	1.0×10^{-8}
Total	5.7×10^{-3}	2.0×10^{-3}
Radionuclide		
Hydrogen-3	5.7×10^{-3}	2.0×10^{-3}
Krypton-85	6.7×10^{-7}	2.4×10^{-7}
Radon-220	2.0×10^{-6}	1.6×10^{-8}
Total	5.7×10^{-3}	2.0×10^{-3}

TABLE 4.19

Radiological Airborne Releases from Building 350, 1999

Direction	Distance to Perimeter (m)	Dose ^a (mrem/yr)	Distance to Nearest Resident (m)	Dose ^a (mrem/yr)
N	1,700	4.7×10^{-4}	2,200	3.3×10^{-4}
NNE	1,800	4.6×10^{-4}	3,200	2.0×10^{-4}
		3.0×10^{-4}	•	1.9×10^{-4}
NE	2,200		3,100	
ENE	2,000	3.4×10^{-4}	3,100	1.8×10^{-4}
E	1,700	5.3×10^{-4}	2,500	2.4×10^{-4}
ESE	900	9.5×10^{-4}	3,000	1.8×10^{-4}
SE	900	6.5×10^{-4}	3,000	1.5×10^{-4}
SSE	700	1.3×10^{-3}	2,700	2.1×10^{-4}
S	600	4.7×10^{-4}	2,700	1.0×10^{-4}
SSW	400	1.7×10^{-3}	2,500	3.3×10^{-4}
SW	600	2.0×10^{-3}	2,700	4.0×10^{-4}
WSW	800	9.6×10^{-4}	2,100	3.0×10^{-4}
W	900	5.8×10^{-4}	2,200	2.5×10^{-4}
WNW	1,000	4.1×10^{-4}	2,100	1.9×10^{-4}
NW	1,900	2.1×10^{-4}	2,400	1.6×10^{-4}
NNW	1,900	2.9×10^{-4}	2,200	2.4×10^{-4}

a Source terms: uranium-234 = 3.2×10^{-5} Ci uranium-238 = 3.2×10^{-5} Ci plutonium-238 = 1.5×10^{-9} Ci plutonium-239 = 5.8×10^{-9} Ci plutonium-240 = 8.5×10^{-9} Ci plutonium-241 = 2.2×10^{-7} Ci plutonium-242 = 1.8×10^{-9} Ci plutonium-244 = 3.3×10^{-13} Ci.

TABLE 4.20

Maximum Perimeter and Individual Doses from Building 350 Air Emissions, 1999 (dose in mrem/yr)

Pathway	Perimeter (600 m SW)	Individual (2,700 m SW)
Ingestion	1.5×10^{-5}	3.1×10^{-6}
Inhalation	2.0×10^{-3}	4.0×10^{-4}
Air immersion	1.2×10^{-12}	2.5×10^{-13}
Ground surface	2.8×10^{-7}	5.6×10^{-8}
Ground surrace	2.0 × 10	3.0 × 10
Total	2.0×10^{-3}	4.0×10^{-4}
Radionuclide		
Uranium-234	1.1×10^{-3}	2.1×10^{-4}
Uranium-238	9.5×10^{-4}	1.9×10^{-4}
Plutonium-238	1.2×10^{-7}	2.4×10^{-8}
Plutonium-239	5.0×10^{-7}	9.9×10^{-8}
Plutonium-240	7.4×10^{-7}	1.5×10^{-7}
Plutonium-241	2.9×10^{-7}	5.8×10^{-8}
Plutonium-242	1.5×10^{-7}	2.9×10^{-8}
Plutonium-244	2.7×10^{-11}	5.3×10^{-12}
Total	2.0×10^{-3}	4.0×10^{-4}

TABLE 4.21

Radiological Airborne Releases from Building 375 (IPNS), 1999

Direction	Distance to Perimeter (m)	Dose ^a (mrem/yr)	Distance to Nearest Resident (m)	Dose ^a (mrem/yr)
N	1,600	5 0 × 10-3	2 200	1.6 × 10-3
N	1,600	5.8×10^{-3}	3,200	1.6×10^{-3}
NNE	1,700	6.1×10^{-3}	3,100	1.8×10^{-3}
NE	1,700	5.4×10^{-3}	2,700	2.1×10^{-3}
ENE	1,500	5.7×10^{-3}	2,500	2.2×10^{-3}
E	600	3.3×10^{-2}	2,500	3.1×10^{-3}
ESE	600	2.6×10^{-2}	2,500	2.2×10^{-3}
SE	600	1.8×10^{-2}	2,500	1.6×10^{-3}
SSE	600	2.6×10^{-2}	3,000	1.5×10^{-3}
S	800	7.5×10^{-3}	3,000	7.7×10^{-4}
SSW	800	2.1×10^{-2}	3,500	1.5×10^{-3}
SW	800	2.8×10^{-2}	4,000	1.4×10^{-3}
WSW	1,500	5.6×10^{-3}	2,700	$1.9\times10^{\text{-3}}$
W	2,200	3.2×10^{-3}	2,700	2.0×10^{-3}
WNW	1,500	4.0×10^{-3}	2,600	1.5×10^{-3}
NW	2,200	1.8×10^{-3}	2,500	1.4×10^{-3}
NNW	1,800	3.2×10^{-3}	2,200	2.2×10^{-3}

^a Source terms: carbon-11=118.1 Ci argon-41 =1.6 Ci.

TABLE 4.22

Maximum Perimeter and Individual Doses from Building 375 (IPNS) Air Emissions, 1999 (dose in mrem/yr)

Pathway	Perimeter (600 m E)	Individual (2,400 m E)
Ingestion	_a	
Ingestion		-
Inhalation	1.4×10^{-3}	1.4×10^{-4}
Air immersion	3.0×10^{-2}	2.8×10^{-3}
Ground surface	1.2×10^{-3}	1.3×10^{-4}
Total	3.3×10^{-2}	3.1×10^{-3}
Radionuclide		
Carbon-11	3.2×10^{-2}	3.1×10^{-3}
Argon-41	5.6×10^{-4}	6.3×10^{-5}
Total	3.3×10^{-2}	3.1×10^{-3}

^a A hyphen indicates no exposure by this pathway.

TABLE 4.23

Radiological Airborne Releases from Building 411/415 (APS), 1999

Direction	Distance to Perimeter (m)	Dose ^a (mrem/yr)	Distance to Nearest Resident (m)	Dose ^a (mrem/yr)
N	1,500	1.6×10^{-4}	2,000	8.5×10^{-5}
NNE	1,600	1.5×10^{-4}	2,100	8.4×10^{-5}
NE	2,200	6.7×10^{-5}	3,100	3.1×10^{-5}
ENE	2,500	4.5×10^{-5}	3,300	2.4×10^{-5}
E	1,600	1.6×10^{-4}	3,400	3.1×10^{-5}
ESE	1,500	1.3×10^{-4}	3,500	2.2×10^{-5}
SE	400	1.2×10^{-3}	3,000	2.0×10^{-5}
SSE	400	1.6×10^{-3}	3,000	2.8×10^{-5}
S	350	8.8×10^{-4}	2,500	2.2×10^{-5}
SSW	400	2.1×10^{-3}	2,800	4.2×10^{-5}
SW	550	1.6×10^{-3}	3,000	4.3×10^{-5}
WSW	800	4.7×10^{-4}	1,400	1.6×10^{-4}
W	800	4.7×10^{-4}	1,500	1.4×10^{-4}
WNW	500	7.3×10^{-4}	1,400	1.1×10^{-4}
NW	350	1.2×10^{-3}	1,600	7.5×10^{-5}
NNW	1,500	1.0×10^{-4}	2,000	5.6×10^{-5}

^a Source terms: carbon-11 = 0.07 Ci (estimated) nitrogen-13 = 3.22 Ci (estimated)

oxygen-15 = 0.35 Ci (estimated).

TABLE 4.24

Maximum Perimeter and Individual Doses from Building 411/415 (APS) Air Emissions, 1999 (dose in mrem/yr)

Pathway	Perimeter (400 m SSW)	Individual (1,400 m WSW)
Ingestion	_a	_
Inhalation	6.1×10^{-5}	$4.6 imes 10^{-6}$
Air immersion	2.0×10^{-3}	1.5×10^{-4}
1 111 1111110101011	3.6×10^{-5}	3.1×10^{-6}
Ground surface	5.0 × 10	3.1 × 10
Total	2.1×10^{-3}	1.6×10^{-4}
Radionuclide		
Carbon-11	4.6×10^{-5}	$4.0 imes 10^{-6}$
Nitrogen-13	1.9×10^{-3}	1.5×10^{-4}
Oxygen-15	1.3×10^{-4}	$4.8\times10^{\text{-}6}$
Total	2.1×10^{-3}	1.6×10^{-4}

^a A hyphen indicates no exposure by this pathway.

The individual doses to the maximally exposed member of the public and the maximum fence line dose are shown in Figure 4.5. The decreases in individual and population doses since 1988 are due in part to the decrease of radon-220 emissions as a result of the cleanup of the Building 200 M-Wing hot cells. The decrease from 1998 to 1999 is principally due to reduced emissions from IPNS because of lower operating time.

The population data in Table 1.1 were used to calculate the cumulative population dose from gaseous radioactive effluents from ANL-E operations. The results are given in Table 4.25, along with the natural external radiation dose. The natural radiation dose listed is the product of the 80-km (50-mi) population and the natural radiation dose of 300 mrem/yr. It is assumed that this dose is representative of the entire area within an 80-km (50-mi) radius. The population dose resulting from ANL-E operations since 1987 is shown in Figure 4.6.

The potential radiation exposures by the inhalation pathways also were calculated by the methodology specified in DOE Order 5400.5. ¹¹ The total quantity for each radionuclide inhaled, in microcuries (μ Ci), is calculated by multiplying the annual average air concentrations by the general public breathing rate of $8,400 \text{ m}^3/\text{yr}$. This annual intake is then multiplied by

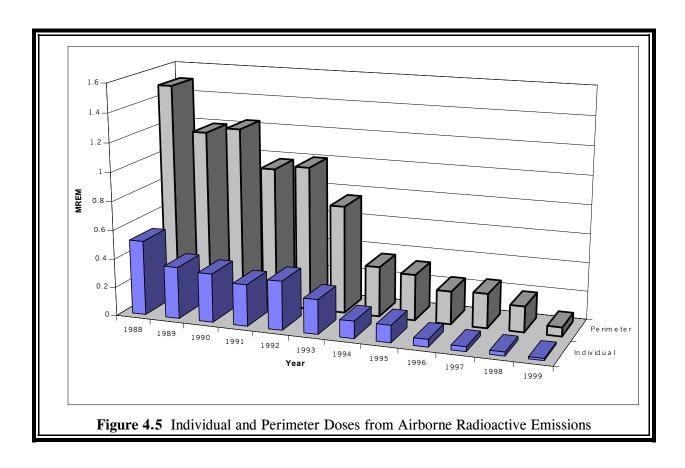
TABLE 4.25

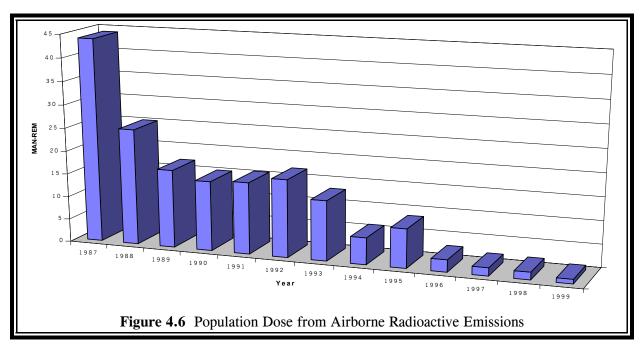
Population Dose within 80 km, 1999

Radionuclide	Man-rem
Hydrogen-3	0.24
Carbon-11	0.15
Nitrogen-13	< 0.01
Oxygen-15	< 0.01
Argon-41	< 0.01
Krypton-85	< 0.01
Thallium-208	< 0.01
Lead-212	0.44
Bismuth-212	0.04
Radon-220	< 0.01
Uranium-234	0.05
Uranium-238	0.04
Plutonium-238	< 0.01
Plutonium-239	< 0.01
Plutonium-240	< 0.01
Plutonium-241	< 0.01
Plutonium-242	< 0.01
Plutonium-244	< 0.01
Total	0.97
Natural	2.5×10^6

the CEDE for the appropriate lung retention class.¹¹ Because the CEDE factors are in units of rem/µCi, this calculation gives the 50-year CEDE. Table 4.26 lists the applicable CEDE factors.

The calculated doses in Tables 4.2 and 4.3 were derived by using this procedure. Because they are all essentially at perimeter locations, these doses represent the fence-line values for those radionuclides measured. In most cases, these doses also are the same as the off-site measurements and represent the ambient dose for the area from these nuclides. No doses were calculated for the total alpha and total beta measurements because the guidance does not provide CEDE factors for such measurements.





4.6.2. Water Pathway

Following the methodology outlined in DOE Order 5400.5, 11 the annual intake of radionuclides (in μ Ci) ingested with water is obtained by multiplying the concentration of radionuclides in microcuries per milliliter (μ Ci/mL) by the average annual water consumption of a member of the general public (7.3 \times 10⁵ mL). This annual intake is then multiplied by the CEDE factor for ingestion (Table 4.26) to obtain the dose received in that year. This procedure was carried out for all radionuclides, and the individual results were summed to obtain the total ingestion dose.

The only significant location where radionuclides attributable to ANL-E operations could be found in off-site water was Sawmill Creek below the wastewater outfall (see Table 4.7. Although this water is not used for drinking purposes, the 50-year effective dose equivalent was calculated for a hypothetical individual ingesting water at the radionuclide concentrations measured at that location. Those radionuclides added to Sawmill Creek by ANL-E wastewater, their net concentrations in the creek, and the corresponding dose rates (if water at these concentrations was used as the sole water supply by an individual) are given in Table 4.27. The dose rates were all well below the standards for the general population. It

50-Year Committed Effective Dose Equivalent (CEDE) Factors

TABLE 4.26

(rem/µCi)

Nuclide	Ingestion	Inhalation
Hadaaaa 2	C 2 10-5	0.610-5
Hydrogen-3	6.3×10^{-5}	9.6×10^{-5}
Beryllium-7	_a	2.7×10^{-4}
Carbon-11	-	8.0×10^{-6}
Strontium-90	0.13	1.32
Cesium-137	0.05	0.032
Lead-210	-	13.2
Radium-226	1.1	-
Thorium-228	-	310
Thorium-230	-	260
Thorium-232	-	1,100
Uranium-234	0.26	130
Uranium-235	0.25	120
Uranium-238	0.23	120
Neptunium-237	3.90	-
Plutonium-238	3.80	-
Plutonium-239	4.30	330
Americium-241	4.50	-
Curium-242	0.11	-
Curium-244	2.30	-
Californium-249	4.60	-
Californium-252	0.94	-

^a A hyphen indicates value not required.

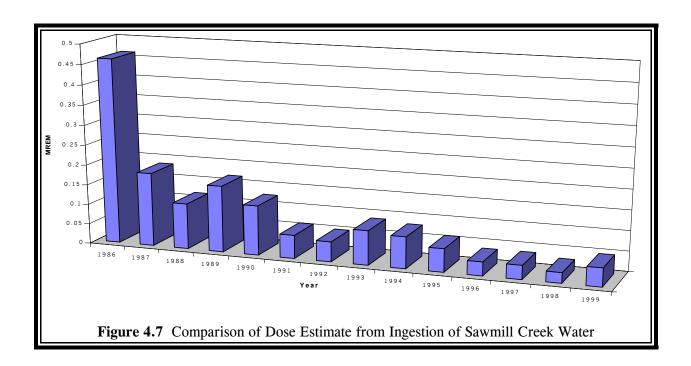
should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows that there are fish in the stream; however, they do not constitute a significant source of food for any individual. Figure 4.7 is a plot of the estimated dose an individual would receive if ingesting Sawmill Creek water.

As indicated in Table 4.7, occasional Sawmill Creek samples (fewer than 10%) contained traces of cesium-137, plutonium-238, curium-242 and 244, or californium-249 and 252; however, the averages were only slightly greater than the detection limit. The annual dose to an individual consuming water at these concentrations can be calculated with the same method used for those

TABLE 4.27

Radionuclide Concentrations and Dose Estimates for Sawmill Creek Water, 1999

Radionuclide	Total Released (Ci)	Net Avg. Concentration (pCi/L)	Dose (mrem)
Hydrogen-3	4.44	370	0.0170
Strontium-90	0.0037	0.31	0.0294
Plutonium-239	0.000001	0.0001	0.00003
Americium-241	0.000005	0.0004	0.00013
Total	4.44		0.0466



radionuclides more commonly found in creek water; this method of averaging, however, probably overestimates the true concentration. Annual doses range from 3×10^{-3} to 6×10^{-5} mrem/yr for these radionuclides.

DOE Order 5400.5¹¹ requires an evaluation of the dose to aquatic organisms from liquid effluents. The dose limit is 1 rad/day or 365 rad/yr. The location that could result in the highest dose to aquatic organisms is in Sawmill Creek downstream of the point where ANL-E discharges its treated wastewater. Inspection of the creek at this location indicates the presence of small bluegill

and carp (about 100 g [4 oz] each). A dose can be estimated by using the annual average concentrations of the radionuclides listed in Table 4.7. The sum of the exposure from these radionuclides is estimated to be about 0.01 rad/yr, which is well within the DOE standard. This value, therefore, demonstrates compliance with that portion of the order.

The EPA has established drinking water standards on the basis of a maximum dose of 4 mrem/yr for man-made beta particle and photon-emitting radio nuclides. The EPA standard is 2×10^4 pCi/L for hydrogen-3 and 8 pCi/L for strontium-90. The net concentrations in Table 4.27 correspond to 1.8% (hydrogen-3) and 3.9% (strontium-90) of the EPA standards. No specific EPA standards exist for the transuranic nuclides.

Sawmill Creek flows into the Des Plaines River. The flow rate of Sawmill Creek (see Section 1.6) is about 0.28 m³/s (10 ft³/s); the flow rate of the Des Plaines River in the vicinity of ANL-E is about 25 m³/s (900 ft³/s). Applying this ratio to the concentration of radionuclides in Sawmill Creek listed in Table 4.27, the dose to a hypothetical individual ingesting water from the Des Plaines River at Lemont would be about 0.0005 mrem/yr. Significant additional dilution occurs further downstream. Very few people, either directly or indirectly, use the Des Plaines River as a source of drinking water. If 100 people used Des Plaines River water at the hypothetical concentration at Lemont, the estimated population dose would be about 10⁻⁵ man-rem.

4.6.3. External Direct Radiation Pathway

The TLD measurements given in Section 4.5 were used to calculate the radiation dose from external sources. Above-normal fence-line doses attributable to ANL-E operations were found at the southern boundary near the Waste Storage Facility (Location 7I).

At Location 7I, the fence-line dose from ANL-E was 105 ± 21 mrem/yr. Approximately 300 m (960 ft) south of the fence line (grid 6I), the measured dose was 88 ± 11 rem/yr, slightly higher than the off-site average (80 ± 4 mrem/yr). No individuals live in this area. The closest residents are about 1.6 km (1 mi) south of the fence line. At this distance, the calculated dose rate from the Waste Storage Facility was 0.001 mrem/yr, if the energy of the radiation were that of a 0.66-MeV cesium-137 gamma-ray, and approximately 0.003 mrem/yr, if the energy were that of a 1.33-MeV cobalt-60 gamma-ray.

At the fence line, where higher doses were measured, the land is wooded and unoccupied. All of these dose calculations are based on full-time, outdoor exposure. Actual exposures to individuals would be substantially less because some of the individuals are indoors (which provides shielding) or away from their dwellings for part of the time. In addition to the permanent resident in the area, occasionally visitors may conduct activities around ANL-E that could result in exposure to radiation from this site. Examples of these activities could be cross-country skiing, horseback

riding, or running in the fire lane next to the perimeter fence. If the individual spent 10 minutes per week adjacent to the 317 Area, the dose would be 0.003 mrem/yr at the 317 Area fence (location 7I) from ANL-E operations.

4.6.4. Dose Summary

The total effective dose equivalent received by off-site residents during 1999 was a combination of the individual doses received through the separate pathways. Radionuclides that contributed through the air pathway are hydrogen-3, carbon-11, nitrogen-13, oxygen-15, argon-41, krypton-85, radon-220 (plus daughters), and actinides. The highest dose was approximately 0.015 mrem/yr to individuals living north of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80-km (50-mi) radius was 0.97 man-rem. The dose pathways are presented in Table 4.28 and compared with the applicable standards.

To receive the maximum public dose, an individual would need to live north of the site at the point of maximum air and direct radiation exposure and use only water from Sawmill Creek below the ANL-E wastewater outfall. This is a very conservative and unlikely situation. To put the maximum individual dose of 0.076 mrem/yr attributable to ANL-E operations into perspective, comparisons can be made with annual average doses received by the public from natural or accepted sources of radiation. These values are listed in Table 4.29. The magnitude of the doses received from ANL-E operations is insignificant compared with these sources. Therefore, the monitoring program results establish that the radioactive emissions from ANL-E are very low and do not endanger the health or safety of those living in the vicinity of the site.

TABLE 4.28

Summary of the Estimated Dose to the Public, 1999

(mrem/yr)

Pathway	ANL-E Estimate	Applicable Standard
Air (less radon)	0.0043	10 (EPA)
Air total	0.0154	100 (DOE)
Water	0.046	100 (DOE)
Direct radiation	0.010	100 (DOE)
Maximum public	0.076	100 (DOE)

TABLE 4.29

Annual Average Dose Equivalent in the U.S. Population^a

Source	Dose (mrem)
Natural	
Radon	200
Internal (potassium-40 and radium-226)	39
Cosmic	28
Terrestrial	28
Medical	
Diagnostic X-rays	39
Nuclear medicine	14
Consumer Products Domestic water supplies, building materials, etc.	10
Occupational (medical radiology, industrial	
radiography, research, etc.)	1
Nuclear fuel cycle	<1
Fallout	<1
Other miscellaneous sources	<1
Total	360

^a National Council on Radiation Protection and Measurements Report No. 93.¹⁸